PROBING SPIN-ORBIT COUPLING OF ORGANOCERIUM RADICALS FORMED IN Ce ATOM REACTIONS WITH ALKYLAMINES.

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Ce atom reactions with alkyamines are carried out in a pulsed-laser ablation molecular beam source and characterized by mass-analyzed threshold ionization (MATI) spectroscopy. The MATI spectra of CeNR (R = CH_3, C_2H_5, and C_3H_7) formed by Ce reactions with H_2NR exhibit two band systems, separated by 78, 74, and 72 cm^{-1}, respectively. In contrast, the MATI spectrum of $CeNC_2H_5$ formed in the Ce + $HN(CH_3)_2$ reaction show two band systems with a much larger separation, 130 cm^{-1}. These separations are attributed to the spin-orbit (SO) splitting from the Ce $4f^1$ electron. The different splittings between CeNR from the reactions of primary amines and $CeNC_2H_5$ from the reaction of secondary amine are due to their different structures. The CeNR complexes from the primary amines have acyclic structures with Ce double bonding to the N atom, whereas $CeNC_2H_5$ from the dimethylamine has a cyclic structure with Ce bonding to the N atom and one of the C atoms. The considerably smaller SO splittings in the CeNR species suggests that N coordination has a stronger quenching effect on the SO coupling of the Ce 4f electron than the C coordination.